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# High Pressure and High Temperature In-situ X-ray Radiography of the Segregation of Molten Iron from Partially Molten Silicate

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## 1 Introduction

X-ray radiography combined with synchrotron radiation provides unique information for studying the behavior of samples at high pressure and high temperature. In geophysical sciences, this technique has been applied, for example, to determine the viscosity of molten silicate [1-3], the interfacial tension of molten iron alloy using the sessile drop method [4, 5], and the total strain of specimens under deformation experiments [6-8].

The Earth's core is believed to have been formed by the segregation of molten iron from the silicate mantle of the undifferentiated proto-Earth. Although much work has been conducted regarding the segregation process, the results were based on indirect evidence, such as textural observations of recovered samples [9,10] by X-ray CT observations [11, 12], or by in-situ electrical resistance measurements [13]. Accordingly, there is a lack of studies based on in situ dynamic observations of the separation process at high pressure and high temperature. Therefore, we have attempted to conduct such observations for the separation of molten iron from silicate using X-ray radiography. Based on these observations, we demonstrate that X-ray radiography is an extremely useful tool for studying such phenomena.

## 2 Experiment

Two types of starting mixtures were prepared to obtain hydrogen-containing and sulfur-containing samples. The hydrogen-containing sample was a mixture of Fe, $SiO_2$, and $Mg(OD)_2$ powder at a molar ratio of 2:1:1. The sulfur-containing sample was a mixture of Fe, FeS, $SiO_2$, and MgO powder at a molar ratio of 0.66:1.17:1:1.

High-pressure and high-temperature in situ X-ray radiography experiments were performed using synchrotron radiation at the AR-NE7 beamline of the PF in KEK. The setup of the X-ray radiography system is summarized in Ref. 14. The radiography images were obtained using an exposure time of 0.5 s/frame, and were recorded to hard disk every 1.5 s. Monochromatic X-rays of 35 keV produced with a double crystal spectrometer (Si(111)) were used to obtain the best absorption image contrast from the sample.

High pressure was generated with a double-stage multi-anvil system using the MAX-III 700 ton press of AR-NE7. Tungsten carbide cubes with a truncated edge length of 12 mm were used as second-stage anvils. The sample was first pressurized to 5 GPa at room temperature and then heated to 1827 K (for the hydrogen-containing samples) or 1973 K (for the sulfur-containing samples) at a rate of approximately 100 K/min while maintaining the load.

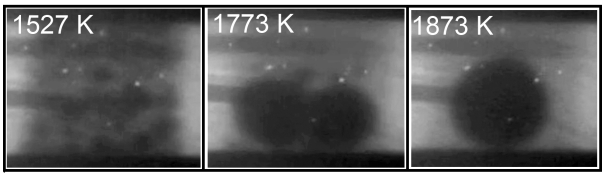

Fig. 1 X-ray radiography images of hydrogen-containing sample observed at 5 GPa with increasing temperature.

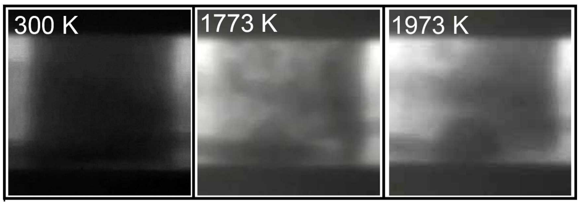

Fig. 2 X-ray radiography images of sulfur-containing sample observed at 5 GPa with increasing temperature.

## 3 Results and Discussion

A series of screenshots of the X-ray radiography images, obtained during heating at 5 GPa from a video of the system, are shown in Figs. 1 and 2. As the temperature of the hydrogen-containing sample increased, cohesion of the molten iron occurred at first and small iron droplets were formed. These iron droplets then sank to the bottom of the sample chamber, where they eventually merged into a single, large molten iron droplet. However, the small iron droplets did not move only vertically down during heating; movement also occurred in random directions. This behavior was markedly different from that observed in the sulfur-containing sample, where the molten iron flowed slowly to the wall of the sample chamber as the temperature of the sample gradually increased. It is thought that such motion may have been driven by a temperature or pressure gradient. These observations demonstrate that the behavior of molten iron changes according to the dissolved elements present.

As demonstrated in the present study, a great deal of information regarding the process of molten iron segregation from partially molten silicate during heating can be obtained using in situ X-ray radiography. This level of information cannot be obtained using quench experiments alone. The combination of such in situ X-ray radiography with tomography observations of the quenched sample will provide rich information about the entire segregation process. This study has also demonstrated that the behavior of molten iron changes dramatically depending on which light element is incorporated into the system. The in situ observation method described here, particularly when combined with recovered experiments or tomography techniques using quench samples, will act as a very useful and powerful tool that will aid future studies of the core formation mechanism of the Earth and other planets.

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